IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:	)	
Tadashi Kokubo et al.	)	
	)	Group Art Unit: 1617
Application No.: 09/936,166	)	
	)	Examiner: Shahnam J. Sharareh
Filed: September 10, 2001	)	
	)	
For: Radioactive Microspheres Excellent	)	
in Chemical Durability and Method	)	
for Manufacturing the Same	)	

DECLARATION

Hon. Commissioner of Patent and Trademarks
Washington, D.C. 20231

Sir:

I, Yoshiaki INOUE, declare and state that:

1. I graduated from Industrial Chemistry department, Faculty of Science and Engineering, Tokyo University of Science in March, 1983. I have been working for Neturen.Co.,Ltd., in Tokyo, Japan, as a research chemist since April 1983 till March 1998, a chief research engineer since April 1998 till March 2004 and a deputy general manager since April 2004 till now, particularly in the field of high frequency induction thermal-plasma technology.

2. I understand that the above application has been rejected over Gray US Patent 5,885,547.

In order to show the differences between the subject matter of Gray Patent and the present invention, the following two experiments were performed under my direction. Experiment A is almost the same as Example 1 of the present specification. Experiment B is a recurrence test of Gray's Example 3.

Experiment A

Yttria powder, approx. size range between 10 to 40 microns and purity of 3N (Shin-Etsu Chemical Co., Ltd.), was melted with a high frequency induction thermal-plasma (ICP No.3 manufactured by Neturen.Co.,Ltd.) under the following condition to form into spheres.

Powder feed carrier gas: Ar 5L/min
Plasma gas composition: Ar 90 L/min + O_2 5L/min
High frequency generator: anode input 40 kW,

output frequency 4 MHz

The spheres were dispersed in ultra pure water with a specific conductance of 18 M? ·cm, and sieved using a 20 μm nylon sieve to remove the fines.

On examination of the surface morphology of the resultant spheres by using the scanning electron microscope (SEM), uniform microspheres having a smooth surface morphology were observed as shown in the following picture. Any spheres having a diameter of more than 41 μm were not observed at all. The tap density of the 20-41 μm microspheres was 3.0 g/cm^3.

Experiment B

Yttria (Y_2O_3) powder, approx. size range between 3 to 10 microns and purity of 4N (Kojundo Chemical Lab. Co.,Ltd), used as starting material was subjected to wet attrition milling to reduce the particle size of the powder to D90=0.9 μm (D50=0.6 μm). 1 kg of milling media (3 mm diameter yttria partially stabilized zirconia spheres obtained from Nikkato Corporation) was placed in a 1L polyethylene pot with up to 100 g of powder.
Sufficient ethanol was added to fill the pot to about 4 mm above the powder and milling media.

The powder was milled (approx. 11 hours) until the size of the majority of the particles was observed to be less than 0.9 micron using a scanning electron microscope (SEM).

After milling, the milling media was separated from the powder using a 0.4 mm sieve. The powder was washed with ethanol, and stored as wet slurry for spray drying.

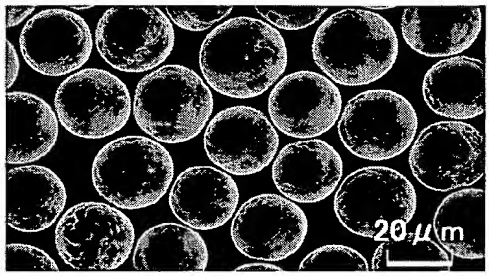
Polyvinyl butyral (PVB, polymerization degree≒700) was added as a binder at a concentration of 8 wt %. The slurry was concentrated to a concentration of 45 wt % using a rotary evaporator (RE52 manufactured by Yamato Scientific Co.,Ltd.), and then spray-dried using a dryer (GS31 manufactured by Yamato Scientific Co.,Ltd.). The inlet temperature (160℃) and the outlet temperature (90±3℃) were monitored by thermocouples.

Prior to plasma spraying, the agglomerates were sieved using a 38 μm nylon sieve to remove the fines while a 100 μm nylon sieve was used to screen out the coarse particles.

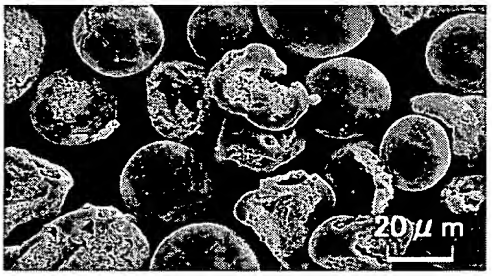
The spray dried powder agglomerates were then plasma sprayed using a DC plasma torch (Plasmaspray SP500S manufactured by Nagata Tekko Co.,Ltd, 50kVA, 400A, 40V).
The plasma gases were Ar and He. The agglomerates were fed into the torch between 4-6 g/min. The plasma sprayed material was deposited on an inner wall of a stainless steel vessel and then collected.

The material was sieved using a 20 μm nylon sieve to remove the fines while a 41 μm nylon sieve was used to screen out the coarse spheres.

On examination of the surface morphology of the resultant 20-41 μm material by using the scanning electron microscope (SEM), irregular microspheres having a rough surface morphology were observed as shown in the following picture. Most of them looked fragmented. The tap density of the 20-41 μm microspheres was 1.3 g/cm^3.



SEM photograph of Experiment A microspheres.



SEM photograph of Experiment B microspheres.

3. I further declare that all statements made herein of my own knowledge are true and that all statements based on information and belief are believed to be true; and further that these statements were made with knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section1001 of Title 18 of the United States Code and that such willful false statements may jeoparadize the validity of the application or any patent issuing thereon.

Date: Aug. 19, 2004

Yoshiaki Inoue

Yoshiaki INOUE